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### TECHNICAL FIELD

[Industrial Application] About a fuel cell, especially this invention is a fuel cell which generates electricity by reforming a fuel, and relates to a fuel cell with little air pollution.

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### DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Industrial Application] About a fuel cell, especially this invention is a fuel cell which generates electricity by reforming a fuel, and relates to a fuel cell with little air pollution. [0002]

[Description of the Prior Art] As a conventional technique, the fuel cell which takes out a current to the exterior is known, using oxygen as an oxidizer, using hydrogen as a reducing agent.

[0003] Drawing 6 is the block diagram showing the conventional fuel cell (single cel) roughly. With reference to drawing 6, this fuel cell 600 contains a negative electrode 608, the electrolyte layer 601, and a positive electrode 609. Through the electrolyte layer 601, a negative electrode 608 and a positive electrode 609 are formed in the center so that phase opposite may be carried out mutually. A negative electrode 608 is formed so that the front face may touch the electrolyte layer 601, and a positive electrode 609 is formed so that the front face may touch the electrolyte layer 601. A negative electrode 608 and a positive electrode 609 are formed by porous members, such as carbon, respectively. Moreover, a negative electrode 608 and a positive electrode 609 include the catalyst of platinum etc., respectively. Moreover, as an electrolyte layer 601, cation exchange membrane is used, for example.

[0004] A negative-electrode active material is supplied to a negative electrode 608 more nearly continuously than the exterior, and positive active material is supplied to a positive electrode 609 more nearly continuously than the exterior.

[0005] As a negative-electrode active material, a fuel (reducing agent) is used and an oxidizer is used as positive active material. Moreover, the resultant generated within the fuel cell 600 is constituted so that it may be removed out of a cel without delay.

[0006] As a fuel (reducing agent), hydrogen is used, for example and oxygen is used as an oxidizer, for example.

[0007] Next, the hydrogen-oxygen system fuel cell which used cation exchange membrane is taken for an example as an electrolyte layer 601, and actuation (generation-of-electrical-energy principle) of a fuel cell 600 is explained below.

[0008] First, the external load circuit 604 which has the external load 605 between a negative electrode 608 and a positive electrode 609 is connected. Then, in a negative electrode 608, the reaction shown below arises, and a hydrogen ion (H+) is generated, and an electron is emitted to the external load circuit 604.

[0009]

[Formula 1] The electron with which oxygen has passed along the external load circuit 604 by the method of H2 ->2H++2e-1, and the positive electrode 609, and H+ which has passed along cation exchange membrane It reacts and water is generated by the reaction shown below. [0010]

[Formula 2] 2H++2e-+1/2O2 ->H2 O -- this time -- between a negative electrode 608 and positive electrodes 609 -- about 1 -- the electromotive force of V is acquired.

[0011] By the way, in the fuel cell of practical use level, in order to raise the generated voltage, there are no dozens in a serial and the above-mentioned fuel cell (single cel) is carried out hundreds laminatings.

[0012] Moreover, when advancing the above-mentioned electrode reaction smoothly, as for the hydrogen used as a fuel, it is desirable that it is pure hydrogen.

[0013] However, in order the price of hydrogen is generally high and to store hydrogen, it is necessary to use a high pressure tank and a hydrogen storing metal alloy. For this reason, hydrogen was stored outside and the fuel cell using the stored hydrogen as a fuel (reducing agent) had the problem that the manufacturing cost and running cost of a fuel cell became high.

[0014] Invention which was indicated by JP,4-129173,A exists as a technique which solves this problem. The fuel cell indicated by JP,4-129173,A is equipped with the generation-of-electrical-energy section which generates electricity using the hydrogen taken out with the reforming machine and reforming machine for taking out hydrogen from natural gas etc. and water.

[0015] Drawing 7 is the sectional view showing roughly the important section of the fuel reforming machine indicated by JP,4–129173,A. With reference to drawing 7, this reforming machine 700 contains the reforming coil 701 and the hydrogen column 702 arranged in the interior of the reforming coil 701. The reforming catalyst 704 of nickel etc. is put in a part of opening section 703 formed by the reforming coil 701 and the hydrogen column 702. Moreover, as for the hydrogen column 702, it comes to form palladium membrane in the metallic conduit of porosity nature. Methane (CH4) and water (H2 O) are supplied to the opening section 703. The reaction shown below arises within the reforming coil 701.

[0016]

[Formula 3] the hydrogen (H2) generated CH4+H2 O->CO+CO2+H2 O+H2 is taken out in the hydrogen column 702 -- having -- moisture (H2 O), and CO and CO2 etc. -- it is emitted to atmospheric air.

[0017] With reference to drawing 6 and drawing 7, the hydrogen taken out in the hydrogen column 702 is sent to a negative electrode 608, and a generation of electrical energy is presented with it. [0018] The fuel cell indicated by JP,4-129173,A has pure hydrogen (H2) obtained from fuels, such as CH4, C two H6, C three H8, and CH3 OH, by the generation-of-electrical-energy section of a fuel cell by performing fuel reforming, and has the advantage in which an efficient generation of electrical energy can be performed.

[0019] Thus, the fuel cell which forms a reforming machine, reforms natural gas, alcohol, etc. with a reforming vessel as equipment for generating hydrogen from natural gas, alcohol, etc. conventionally for the purpose of reducing the manufacturing cost and running cost of a fuel cell, generates hydrogen, and generates electricity using the generated hydrogen has been proposed variously. [0020] the hydrogen generating approach performed in the conventional reforming machine — material gas — a steam — in addition, the steam reforming process which transforms coal—for—coke—making—ized hydrogen to hydrogen, a carbon monoxide, and a carbon dioxide and the partial oxidation (partial combustion) which a part of coal—for—coke—making—ized hydrogen is burned and obtains hydrogen, a carbon monoxide, and a carbon dioxide — there is law.

[0021] Generally it is Cn Hm about a hydrocarbon. If expressed, the reaction which occurs in a steam reforming process can be shown as a reaction shown below.

[0022]

[Formula 4]

Cn Hm+nH2 O->nCO+(n+m/2) H2 -- again -- the same -- a hydrocarbon -- general -- Cn Hm If expressed, the reaction which occurs in a partial oxidation method can be shown as a reaction shown below.

[0023]

[Formula 5] Cn Hm+n/2O2 ->nCO+m/2H2 -- in these reactions, liftings and these reactions maintain a balance and advance the reaction which shows some carbon monoxides below. [0024]

[Formula 6] CO+H2 O->CO2+H2[0025]

[Formula 7] The reaction shown in the CO+3H2 ->CH4+H2 O-ization 4 is endothermic reaction, and other reactions are exothermic reaction. Therefore, it is necessary to heat from the outside and a reaction is usually performed in the temperature of 800 degrees C - 900 degrees C at the reaction shown in \*\* 4 by the coil of heat-resistant metal filled up with the catalyst of a nickel system. On the other hand at such temperature, the reaction of \*\* 6 and \*\* 7 seldom advances on right-hand side, for example, in the case of a 850-degree C reaction, the concentration of a carbon dioxide is 13% - 14%, and the concentration of methane is 3% - 4%.

[0026] Moreover, when the methanol which is a kind of alcohol instead of a hydrocarbon is used, reforming advances at a reaction like water and a degree type.
[0027]

[Formula 8] CH3 OH+nH2 O->(1-n) CO+nH2 O+nGO2+(2+n) H2, however the reaction shown in the O<n<1-ization 8 are 0kg/cm2 - 20kg/cm2. It is made desirable to set it as a pressure and the temperature of 200 degrees C - 600 degrees C. [0028]

[Problem(s) to be Solved by the Invention] After performing the reaction mentioned above, the carbon monoxide of remarkable concentration is contained in the gas which comes out of a reforming machine.

[0029] For this reason, the carbon monoxide contained in the gas which comes out of a reforming machine may carry out poisoning of the catalyst of a fuel cell etc., and may bar a normal cell reaction.

[0030] Then, equipment called a carbon monoxide convertor is used in order to make the amount of survival of a carbon monoxide low, and to push on the right the reaction shown in \*\* 6. In order to push the reaction shown in \*\* 6 which is exothermic reaction, a reaction temperature setup is carried out first at 350 degrees C - about 370 degrees C, in order to raise a reaction rate, the so-called elevated-temperature inversion (Hot Shift) is performed using catalysts, such as an iron-chrome system, and low-temperature inversion (Cold Shift) is made to perform at 200 degrees C - about 230 degrees C further by the carbon monoxide convertor using the catalyst of a copper-zinc system. Thereby, carbon monoxide concentration can be reduced.

[0031] On the other hand, after this reforming reaction, when a fuel is alcohol, in order to reduce CO of reformed gas further, CO shift catalyst is used. As a CO shift catalyst, the catalyst of an iron-chrome system, the catalyst of a copper-zinc system, etc. are used. In CO shift processing, the concentration of CO is reduced for CO by even about 1% of concentration by the reaction with H2 O. Furthermore, in order to reduce CO concentration, it is possible by making reformed gas react with air further with 2nd CO reduction equipment to reduce CO concentration to 100 ppm. However, by such approach, equipment itself becomes complicated and there is a problem of requiring an elevated temperature etc. Especially when using such a fuel cell for an electric vehicle etc., there is a problem that a system becomes large.

[0032] This invention aims at offering the compact fuel cell which can generate electricity using the hydrogen gas which generated hydrogen gas and was generated from a fuel, without almost generating a carbon monoxide (CO), without being made in order to solve the above problems, and using the above complicated flow systems.

[0033]

[0034]

[0035]

[Means for Solving the Problem] The 1st electrolyte layer which the fuel cell according to this invention is formed so that the front face of the 1st electrode which consists of an ingredient

including a catalyst, and the 1st electrode may be touched, and has cation—exchange ability. The 2nd electrode which consists of an ingredient which is prepared so that the front face of the 1st electrolyte layer may be touched, and includes a catalyst, The 3rd electrode which turns into the 1st electrode from the fuel—supply means for supplying a fuel, and an ingredient including a catalyst, The 2nd electrolyte layer which is prepared so that the front face of the 3rd electrode may be touched, and has cation—exchange ability. The 4th electrode which consists of an ingredient which is prepared so that the front face of the 2nd electrolyte layer may be touched, and includes a catalyst, It has a means to supply the reducing agent generated in the 2nd electrode to the 3rd electrode, a means established between the 2nd electrode and the 3rd electrode to energize a current, and an oxidizer supply means for supplying an oxidizer to the 4th electrode, and a current is taken out from the 1st and 4th electrodes.

[0036] The fuel cell according to this invention is formed by the conductive porosity member with same means established between a means to supply preferably the reducing agent generated in the 2nd electrode to the 3rd electrode, and the 2nd electrode and the 3rd electrode to energize a current.

[0037] In addition A porosity member contains the free passage pore which opens the front face of one side, and the front face of the other side for free passage.

[0038] Moreover, the fuel cell according to this invention is desirable, and, in a fuel, a reducing agent contains hydrogen at least, including a methanol and water.

[0039] In addition, the vocabulary "an electrode" used on these specifications contains the electrode constituted by the combination of the electrode for fuels, a counterelectrode, the electrode for fuels, and a separator, and the electrode constituted by the combination of a counterelectrode and a separator.

[0040]

[Function] The mechanism of action of this invention is explained below using drawing.
[0041] <u>Drawing 2</u> is a block diagram which is used in the fuel cell according to this invention and in which showing typically an example of the reducing-agent generation means for disassembling a fuel electrochemically and generating a reducing agent. With reference to <u>drawing 2</u>, this reducing-agent

generation equipment 200 contains the 1st electrode 208, the electrolyte layer 201 which has cation-exchange ability, and the 2nd electrode 209.

[0042] As an electrolyte layer 201 which has cation-exchange ability, the cation exchange membrane which consists of a solid-state polyelectrolyte is used, for example.

[0043] The 1st electrode 208 contains the electrode 202 for fuels of the porosity nature which made the platinum catalyst support, and the conductive separator 206.

[0044] Moreover, the 2nd electrode 209 contains the counterelectrode 203 of the porosity nature which made the platinum catalyst support, and the conductive separator 207.

[0045] Surface 202a of one side of the electrode 202 for fuels is prepared so that surface 201a of one side of the electrolyte layer 201 may be touched.

[0046] Surface 203a of one side of a counterelectrode 203 is prepared so that surface 201b of the other side of the electrolyte layer 201 may be touched.

[0047] The zygote together put so that the electrode 202 for fuels and a counterelectrode 203 might face in the center through the electrolyte layer 201 is pinched with a separator 206 and a separator 207.

[0048] -cum- CO2 for two or more fuel supply as a means for supplying a fuel to field 206a of the side which touches the electrode 202 for fuels of a separator 206, and discharging a resultant 206h of slots for discharge is formed.

[0049] 207h of two or more slots for hydrogen (H2) discharge is formed in field 207a of the side which touches the counterelectrode 203 of a separator 207.

[0050] A fuel is supplied to the electrode 202 for fuels from the exterior. An external circuit 204 is electrically connected to the electrode 202 for fuels, and a counterelectrode 203 through a

separator 206 and a separator 207, respectively.

[0051] DC power supply 205 are formed in the external circuit 204 as a means for impressing an electrical potential difference so that the 1st electrode 208 may be used as a positive electrode and the 2nd electrode 209 may be used as a negative electrode. The electrode 202 for fuels is used as a positive electrode, and let a counterelectrode 203 be a negative electrode.

[0052] In the reducing-agent generation equipment 200 mentioned above, water or a steam is supplied to the electrode 202 for fuels with the methanol which is a fuel, and through an external circuit 204, an electrical potential difference is impressed so that an electron may be drawn out from the electrode 202 for fuels. Consequently, the next reaction advances in the electrode 202 for fuels.

[0053]

[Formula 9]

CH3 OH+2H2 O->CO -- the hydrogen ion generated with the electrode 202 for 2+6e-+6H+ fuels passes cation exchange membrane, and is changed as follows with a counterelectrode 203. [0054]

[Formula 10] 6H++6e-->3H2 -- such a process can be made to generate hydrogen alternatively by the counterelectrode 203 side Moreover, at this reaction, since CO is hardly generated, carbon monoxide concentration can be maintained at a very low level, so that clearly from the reaction formula shown in \*\* 9.

[0055] Next, the mechanism of action of the generation-of-electrical-energy means which generates electricity using the reducing agent generated by the reducing-agent generation means is explained below using drawing.

[0056] <u>Drawing 3</u> is a block diagram which is used in the fuel cell according to this invention and in which showing typically an example of the generation-of-electrical-energy means which generates electricity using the reducing agent generated by the reducing-agent generation means. With reference to <u>drawing 3</u>, this power plant 300 contains the 3rd electrode 308, the electrolyte layer 301 which has cation-exchange ability, and the 4th electrode 309.

[0057] As an electrolyte layer 301 which has cation-exchange ability, the cation exchange membrane which consists of a solid-state polyelectrolyte is used, for example.

[0058] The 3rd electrode 308 contains the electrode 302 for fuels of the porosity generation which made the platinum catalyst support, and the conductive separator 306.

[0059] Moreover, the 4th electrode 309 contains the counterelectrode 303 of the porosity nature which made the platinum catalyst support, and the conductive separator 307.

[0060] Surface 302a of one side of the electrode 302 for fuels is prepared so that surface 301a of one side of the electrolyte layer 301 may be touched.

[0061] Surface 303a of one side of a counterelectrode 303 is prepared so that surface 301b of the other side of the electrolyte layer 301 may be touched.

[0062] The zygote together put so that the electrode 302 for fuels and a counterelectrode 303 might face in the center through the electrolyte layer 301 is pinched with a separator 306 and a separator 307.

[0063] Two or more 306h is formed in field 306a of the side which touches the electrode 302 for fuels of a separator 306 as a means for supplying a fuel (H2).

[0064] An oxidizer is supplied to field 307a of the side which touches the counterelectrode 303 of a separator 307, and 307h of two or more slots for -cum- H2 O discharge for oxygen supply is formed in it as a means for discharging a resultant.

[0065] The hydrogen generated with the reducing-agent generation equipment 200 mentioned above is supplied to the electrode 302 for fuels. Moreover, oxygen (O2) is supplied to a counterelectrode 303 from the exterior.

[0066] The external load circuit 304 which has the external load 305 is connected to the electrode 302 for fuels, and a counterelectrode 303 through a separator 306 and a separator 307, respectively.

Then, in the electrode 302 for fuels, a current flows so that an electron may be drawn out from the electrode 302 for fuels, and the next reaction advances with the current 302 for fuels. [0067]

[Formula 11] The next reaction advances in the method of H2 ->2H++2e-1, and a counterelectrode 303.

[0068]

[Formula 12] 2H++2e-+1/2H2 O->H2 O -- at this time, the fuel electrode 302 can turn into a negative electrode, a counterelectrode 303 turns into a positive electrode, and a current can be taken out outside.

[0069]

[Example] Hereafter, although this invention is explained using an example, this invention is not limited at all depending on the following examples.

[0070] Example 1 drawing 1 is the block diagram showing one example of a fuel cell typically for reference.

[0071] With reference to <u>drawing 1</u>, this fuel cell 100 contains reducing-agent generation equipment 200 and a power plant 300.

[0072] Since reducing-agent generation equipment 200 is the same as the reducing-agent generation equipment 200 shown in <u>drawing 2</u>, the explanation is omitted.

[0073] Moreover, since the power plant 300 is the same as the power plant 300 shown in  $\frac{\text{drawing }3}{\text{drawing }3}$ , the explanation is omitted.

[0074] With reference to <u>drawing 1</u>, <u>drawing 2</u>, and <u>drawing 3</u>, the reducing agent generated with the counterelectrode 203 is supplied to the electrode 302 for fuels by the duct 250 for reducingagent supply.

[0075] In this example, the electrode 202 for fuels which constitutes reducing-agent generation equipment 200, and the counterelectrode 203 were formed as follows.

[0076] First, the cation exchange membrane (trade name NAFION117R (Du Pont make)) which consists of solid-state polyelectrolyte film was prepared as an electrolyte layer 201.

[0077] Next, it is 1%NaBH4 as 3% chloroplatinic acid liquid and a reducing agent as a metal salt. After making a reducing agent permeate cation exchange membrane using a solution, the front face of cation exchange membrane was contacted in the chloroplatinic acid solution, and the platinum layer was deposited. By this approach, on the surface of cation exchange membrane, the platinum catalyst layer of porosity nature was formed and it considered as one pair of electrodes.

[0078] Moreover, in this example, the electrode 302 for fuels which constitutes a power plant 300, and the counterelectrode 303 were formed as follows.

[0079] As a base material of the electrode 302 for fuels, and a counterelectrode 303, the base material of porosity nature which consists of conductive carbon:PTFE(polytetrafluoroethylene) =3:7 was prepared. Next, it is platinum as a catalyst to this base material 5 mg/cm2 It supported both in quality and in quantity. As an electrolyte layer 301, the cation exchange membrane (trade name NAFION117R (Du Pont make)) which consists of solid-state polyelectrolyte film was used. [0080] Next, the mole ratio supplied the methanol-water mixed liquor of methanol:water =1:2, and set the temperature of the cel which constitutes reducing-agent generation equipment 200 as 30 degrees C at the electrode 202 side for fuels which constitutes the reducing-agent generation equipment 200 of this fuel cell 100. And when the electrode 202 side for fuels was considered as plus and the electrical potential difference of 0.2V was applied between the electrode 202 for fuels, and the counterelectrode 203, it is 1.0 mA/cm2. A current flows and it is CO2 from the electrode 202 for fuels. Hydrogen gas was generated from the counterelectrode 203. The carbon monoxide was not detected in the generated hydrogen.

[0081] Next, the hydrogen generated from reducing-agent generation equipment 200 was supplied to the electrode 302 for fuels which constitutes a power plant 300 using the duct 250 for reducing-agent supply.

[0082] When the hydrogen generated by reducing-agent generation equipment 200 was supplied to the electrode 302 for fuels and oxygen was supplied to the counterelectrode 303, in the power plant 200, 1.0 mA/cm2 and the output voltage of 0.9V were obtained.

[0083] Consequently, 1 mA/cm2 When electrical-potential-difference 0.2V needed with reducing-agent generation equipment 200 with the current value were deducted from output voltage 0.9V outputted in the power plant 300, in the fuel cell 100 whole, 1 mA/cm2 and the output voltage of 0.7V were able to be obtained.

[0084] In addition, although this example showed the example which established the external power 205 to reducing-agent generation equipment 200, a power plant 300 may be used as such an external power 205.

[0085] Example 2 drawing 4 is the block diagram showing typically one example of the fuel cell according to this invention.

[0086] With reference to drawing 4, a laminating is carried out one by one and this fuel cell 400 becomes so that a separator 421, the porous current collection layer 406, the electrode 402 for fuels (methanol pole), the cation-exchange-membrane layer 401, a counterelectrode (hydrogen generating pole) 403, the porous current collection layer 420, the electrode 412 for fuels (hydrogen pole), the cation-exchange-membrane layer 411, a counterelectrode (oxygen pole) 413, the porous current collection layer 417, and a separator 422 may touch this order mutually.

[0087] The terminals 423 and 424 for connecting with an external load circuit are formed in a separator 421 and a separator 422, respectively.

[0088] A fuel is supplied to field 406a of the side which touches the electrode 402 for fuels of the porous current collection layer 406, and 406h of two or more slots for -cum- CO2 discharge for fuel supply is formed in it as a means for discharging a resultant.

[0089] the object for two or more hydrogen [ a / of the side which touches the counterelectrode 403 of the porous current collection layer 420 / field 420] discharge -- slot 420ha is formed.

[0090] the object for two or more hydrogen [ b / of the side which touches the electrode 412 for fuels of the porous current collection layer 420 / field 420] supply -- slot 420hb is formed.

[0091] An oxidizer is supplied to field 417a of the side which touches the counterelectrode 413 of the porous current collection layer 417, and 417h of two or more slots for -cum- H2 O discharge for oxygen supply is formed in it as a means for discharging a resultant.

[0092] Moreover, the fuel-supply means (not shown) for supplying a fuel is formed in the electrode 402 for fuels.

[0093] Moreover, the oxidizer supply means (not shown) for supplying an oxidizer is formed in a counterelectrode 413.

[0094] The porous current collection layer 420 consists of members of the porosity nature which has the free passage pore which opens field 420a of one side of the porous current collection layer 420, and field 420b of the other side for free passage.

[0095] Moreover, the means (not shown) for discharging the oxidizer of the surplus supplied to the resultant (H2 O) and counterelectrode 413 which were generated with the counterelectrode 413 from a fuel cell 400 is formed in a counterelectrode 413.

[0096] Next, the case where a methanol and water are used is taken for an example as a fuel, and actuation of a fuel cell 400 is explained below.

[0097] First, an electrical potential difference is impressed so that a methanol and water may be introduced into the electrode 402 for fuels, the electrode 402 for fuels may be used as a positive electrode and a counterelectrode 403 may be used as a negative electrode.

[0098] Then, the reaction of a degree type advances in the electrode 402 for fuels. [0099]

[Formula 13]

CH3 OH+2H2 O->CO2+6H++6e - H+ produced at this reaction It passes along cation exchange membrane 401, and is H2 like a degree type with a counterelectrode 403. It generates.

## [0100]

[Formula 14] The hydrogen generated 6H++6e-->3H2 passes along the free passage pore of the current collection layer 420, and reaches the electrode 412 for fuels (hydrogen pole).

[0101] Next, oxygen is supplied to a counterelectrode (oxygen pole) 413. Moreover, the power source which was being impressed to the electrode 402 for fuels and the counterelectrode 403 is removed. Next, an external load circuit (not shown) is connected to a terminal 423 and a terminal 424.

[0102] Then, in the electrode 412 for fuels (hydrogen pole), an electron is drawn out and a reaction like a degree type is caused.

[0103]

[Formula 15] 3H2 ->6H++6e - H+ produced at this reaction It passes along cation exchange membrane 411, and a reaction like a degree type is caused with a counterelectrode (oxygen pole) 413.

[0104]

[Formula 16] 6H++6e- a current can be continuously taken out outside by supplying a methanol and water to the electrode 402 for fuels continuously, and supplying oxygen to a counterelectrode (oxygen pole) 417 continuously below +2 / 302 ->3H2 O.

[0105] Hereafter, based on concrete data, it explains in more detail. The porosity carbon plate was used as the current collection layer 406, the current collection layer 420, and a current collection layer 417.

[0106] In this example, the electrode 402 for fuels and the counterelectrode (hydrogen generating pole) 403 were formed as follows.

[0107] First, the cation exchange membrane (trade name NAFION117R (Du Pont make)) which consists of solid-state polyelectrolyte film was prepared as a cation-exchange-membrane layer 401. Next, it is 1%NaBH4 as 3% chloroplatinic acid liquid and a reducing agent as a metal salt. After making a reducing agent permeate cation exchange membrane using a solution, the front face of cation exchange membrane was contacted in the chloroplatinic acid solution, and the platinum layer was deposited. By this approach, the porous platinum catalyst layer was formed in both sides of cation exchange membrane, and it considered as one pair of electrodes.

[0108] Moreover, in this example, the electrode 412 for fuels (hydrogen pole) and the counterelectrode (oxygen pole) 413 were formed as follows.

[0109] As a base material of the electrode 412 for fuels, and a counterelectrode 413, the base material of porosity nature which consists of conductive carbon:PTFE(polytetrafluoroethylene) =3:7 was prepared. Next, it is platinum as a catalyst to this base material 5 mg/cm2 It supported both in quality and in quantity.

[0110] As a cation-exchange-membrane layer 411, the cation exchange membrane (trade name NAFION117R (Du Pont make)) which consists of solid-state polyelectrolyte film was used.
[0111] Next, the mole ratio supplied the methanol-water mixed liquor of methanol:water =1:1, and set the temperature of a cel as 30 degrees C at the electrode 402 side for fuels. And when the electrode 402 side for fuels was considered as plus and the electrical potential difference of 0.2V was applied between the electrode 402 for fuels, and the counterelectrode 403, it is 1.0 mA/cm2. A current flows and it is CO2 from the electrode 402 for fuels. Hydrogen gas was generated from the counterelectrode 403. The carbon monoxide was not detected in the generated hydrogen. After continuing this actuation fixed time, oxygen was supplied to the counterelectrode (oxygen pole) 413. Next, the electrical potential difference which was being impressed between the electrode 402 for fuels and the counterelectrode 403 was removed.

[0112] Next, the terminal 423 and the terminal 424 were connected to the external load circuit (not shown). It is 1.0 mA/cm2 at the fuel cell 400 whole by supplying a methanol and water to the electrode 402 for fuels continuously, and supplying oxygen to a counterelectrode (oxygen pole) 413 continuously hereafter. Current density and the output voltage of 0.7V were able to be taken out

continuously.

[0113] In addition, in this example, as the current collection layer 406 and a current collection layer 417, although the base material of porosity nature was used, the current collection layer 406 and the current collection layer 417 may be formed with the precise ingredient.

[0114] Moreover, when a precise ingredient is used for the current collection layer 406 and the current collection layer 417, the separator 421 and the separator 422 write in addition that it is not a required member especially.

[0115] moreover, slot 420ha for hydrogen discharge prepared in the current collection layer 420 and the object for hydrogen supply — slot 420hb writes in addition that it is not the indispensable component of this invention that what is necessary is to just be formed of each and the need. [0116] In addition, a methanol and water are introduced into the electrode 402 for fuels in this example. Although the example which started the fuel cell 400 using the hydrogen which impressed the electrical potential difference so that the electrode 402 for fuels might be used as a positive electrode and a counterelectrode 403 might be used as a negative electrode, was generated by after an appropriate time in the counterelectrode 403, and reached the electrode 412 for fuels, and the oxygen supplied to the counterelectrode 413 was shown The starting approach of a fuel cell 400 is not limited to the above—mentioned starting approach. Hydrogen may be supplied to the electrode 412 for fuels from the exterior, oxygen may be supplied to a counterelectrode 413, and the electrode 402 for fuels may be made to start a fuel cell 400 also by introducing a methanol and water after an appropriate time.

[0117] The fuel cell 400 shown in example 3 example 2 was made into the single cel unit, 4 cel laminating of this single cel unit was carried out to the serial, and the same actuation as an example 2 was performed.

[0118] By the above actuation, current density 1.0 mA/cm2 and output voltage 2.8V were able to be continuously taken out with 4 cel tandem-type fuel cell shown in an example 3.

[0119] In addition, it does not pass over the indication about the above example in the mere example of this invention, and it does not restrict the technical range of this invention at all. [0120] As an electrolyte layer which is used in this invention and which has cation—exchange ability, especially if a cation is made to penetrate alternatively, it will not be limited. The electrolyte layer which has such cation—exchange ability may be a solid—state, or may be liquids, such as the electrolytic solution. As an electrolyte layer which has cation—exchange ability, when using cation exchange membrane, the solid—state polyelectrolyte film, the film which consists of a matrix containing a phosphoric acid, the film which consists of a matrix containing a sulfuric acid, the film which consists of a solid electrolyte can be mentioned. Moreover, the ion exchange membrane which has a sulfonic acid, phosphonic acid, a sulfate, phosphoric ester, etc. can be used as a cation—exchange radical.

[0121] Moreover, in the following cases, especially the catalyst used in this invention is not limited; but it can mention alloys, such as platinum (Pt), a rhodium (Rh), palladium (Pd), a ruthenium (Ru), gold (Au), iridium (Ir) or these metals, and nickel (nickel), cobalt (Co), etc. Moreover, an electrode may be formed from the catalyst ingredient itself, such as an alloy of platinum (Pt), a rhodium (Rh), palladium (Pd), a ruthenium (Ru), gold (Au), iridium (Ir), and these elements which contains either at least, and may be formed from the thing which made the conductive carbon electrode etc. support a catalyst ingredient.

[0122] The electrode according to this invention can be formed by depositing an electrode material in the porous structure of the front face of the film of cation exchange membrane. In addition, the electrode according to this invention can be formed by supporting a catalyst to the porous body beforehand produced by \*\*\*\* etc.

[0123] In this invention, although a fuel-supply means can be equipped with the pump for sending a fuel into the container or tub which holds a fuel, this container, or a tub by the predetermined pressure etc. since it contacts a fuel to one side of an electrode, it is not limited to these, but if it is

the device which supplies a fuel to an electrode, anythings can be used.

[0124] As a fuel used in the fuel cell according to this invention, what contains a methanol and water at least can be mentioned.

[01.25] Moreover, high-voltage-ization also becomes possible by carrying out the laminating of many units shown in an example 2 to a serial. <u>Drawing 5</u> is a block diagram in which such a fuel cell 400 shows typically the fuel cell by which the laminating was carried out to the serial.

[0126] With reference to drawing 5, the laminating of many fuel cells 400 is carried out to a serial, and the generation-of-electrical-energy section of a fuel cell is constituted.

[0127] And a terminal is prepared in the current collection layer of the side to which the fuel of the cell cell unit of the both ends of the fuel cell 400 which comes to carry out a laminating to an a large number serial is supplied, and the current collection layer of the side to which an oxidizer is supplied, respectively, and the external terminal 504 which has the external load 505 is connected to them.

[0128]

[Effect of the Invention] As explained above, according to this invention, the complicated system of reaction and the complicated flow system like before are not required, and can generate hydrogen with high purity with very simple equipment. According to this invention, generation of impurities, such as CO, is controlled.

[0129] For this reason, since CO by which the fuel cell according to this invention is generated is reduced remarkably, the fall of the chemical reactivity by poisoning of a catalyst is also prevented. [0130] Moreover, high-voltage-ization also becomes possible by carrying out the laminating of the fuel cell (single cel) according to this invention to two or more serials.

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### **CLAIMS**

(57) [Claim(s)]

[Claim 1] The 1st electrode which consists of an ingredient including a catalyst, and the 1st electrolyte layer which is prepared so that the front face of said 1st electrode may be touched, and has cation—exchange ability, The 2nd electrode which consists of an ingredient which is prepared so that the front face of said 1st electrolyte layer may be touched, and includes a catalyst, The 3rd electrode which turns into said 1st electrode from the fuel—supply means for supplying a fuel, and an ingredient including a catalyst, The 2nd electrolyte layer which is prepared so that the front face of said 3rd electrode may be touched, and has cation—exchange ability, The 4th electrode which consists of an ingredient which is prepared so that the front face of said 2nd electrolyte layer may be touched, and includes a catalyst, A means to supply the reducing agent generated in said 2nd electrode to said 3rd electrode. The fuel cell which is equipped with a means established between said 2nd electrode and said 3rd electrode to energize a current, and the oxidizer supply means for supplying an oxidizer to said 4th electrode, and takes out a current from said 1st and 4th electrodes.

[Claim 2] The fuel cell according to claim 1 with which a means established between a means to supply the reducing agent generated in said 2nd electrode to said 3rd electrode, and said 2nd electrode and said 3rd electrode to energize a current is formed by the same conductive porosity member.

[Claim 3] For said reducing agent, said fuel is a fuel cell containing hydrogen according to claim 1 or 2 at least, including a methanol and water.

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### **DESCRIPTION OF DRAWINGS**

[Brief Description of the Drawings]

[Drawing 1] It is the block diagram showing typically one example of the fuel cell mentioned to reference.

[Drawing 2] It is the block diagram showing typically one example of the reducing-agent generation equipment of a fuel cell of following this invention.

[Drawing 3] It is the block diagram showing typically one example of the power plant of the fuel cell according to this invention.

[Drawing 4] It is the block diagram showing typically one example of a fuel cell of following this invention.

[Drawing 5] The fuel cell (single cel) according to this invention is the block diagram showing typically one example of the fuel cell by which two or more laminatings were carried out to the serial.

[Drawing 6] It is the block diagram showing the conventional fuel cell typically.

[Drawing 7] It is the block diagram showing the conventional fuel reforming machine typically.

[Description of Notations]

100,400,500 Fuel cell

200 Reducing-Agent Generation Equipment

201, 301, 401, 411 Electrolyte layer which has cation-exchange ability

202, 302, 402, 412 Electrode for fuels

203, 303, 403, 413 Counterelectrode

204, 304, 504 External circuit

205 Power Source

206, 207, 306, 307, 421, 422 Separator

206h, 406h -cum- CO2 for fuel supply Slot for discharge

207h, 420ha Hydrogen Slot for discharge

208, 209, 308, 309 Electrode

300 Power Plant

305 505 External load

306h, 420hb Slot for hydrogen supply

307h, 417h Oxygen Slot for -cum- H2 O discharge for supply

406, 420, 417 Current collection layer

423 424 Terminal

Drawing selection drawing 1

